LA-UR -92-1230

LA-UR--92-1230

DE92 013546

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36

TITLE **MULTIPHOTON PROCESSESS IN SCATTERING AND IONIZATION IN AN INTENSE LINEARLY-POLARIZED RADIATION FIELD**

AUTHOR(S) **G. CSANAK & L.A. COLLINS**

SUBMITTED TO Physics of High Power Laser Matter Interactions
Kyoto, Japan
March 9-13, 1992

DISCLAIMER

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

MULTIPHOTON PROCESSES IN SCATTERING AND IONIZATION IN AN INTENSE LINEARLY–POLARIZED RADIATION FIELD

BY G. CSANAK[+] and L.A. COLLINS
Group T–4, Theoretical Division, Los Alamos National Laboratory
Los Alamos, NM 87545, USA
[+] University of Tsukuba, Tsukuba, Japan

We have employed the Gavrila–Kaminski formulation for the conversion of the time–dependent Schrodinger equation to a set of coupled differential equations that describe the interaction of an electron with a proton in the presence of an intense linearly–polarized radiation field in the Kramers–Henneberger gauge. The differential equations are solved numerically by a linear algebraic prescription effectively applied to electron–molecule collisions. We studied both electron–proton collisons assisted by the laser field and multiphoton ionization of H. In the former, we observed the capture–escape resonances while in the latter, we found evidence of the stabilization mechanism as the intensity reaches 10^{18} W/cm^2.

1. Introduction

The interaction of an intense radiation field with atoms is usually characterized in terms of the time–dependent(TD) Schrcdinger equation, whose solution is complicated by having both spatial and temporal dimensions. Gavrila and Kimenski(1984) proposed a procedure for transforming to a time–independent set of coupled differential equations for the Fourier components of the TD wavefunction in the Kramers–Henneberger gauge(Kramers,1956; Henneberger,1968). In its simplest form, the formulation describes the interaction of an electron with an atomic potential in the presence of a laser field. The technique has been applied to a wide variety of field–assisted processes including scattering, bound–bound transitions, and photoionization(Faisal,1973; Gersten and Mittleman,1976; Guisti–Suzor and Zoller,1989; Pont et.al.,1988 and 1990; Pont,1989; Pont and Gavrila,1990). The method relies upon only a few general hypotheses: 1) the dipole approximation, 2) a nonrelativistic treatment, and 3) a monochromatic radiation field, and within these limitations, provides a completely general approach provided that the coupled equations can be efficiently converged. Most applications to date have dealt with uncoupled systems of equations that represent the high–frequency and high–intensity limit. Such calculations depict actual physical situations only in very limited circumstances. Several groups have extended the calculations to energetically coupled channels(Dimou and Faisal,1984; Franz et.al.,1990; Marte and Zoller,1991) but have restricted the treatment to circularly polarized light and to one–dimensional models(Bhatt et.al.,1988). In addition, Marte and Zoller have developed a multi–channel quantum defect scheme for handling the highly–excited Rydberg–like channels. Limiting applications to circularly polarized light may impose serious restrictions on the technique due to the limited coupling range and the specialized reduction inherent in the rotating frame. In order to demonstrate the full power of the technique, we have performed calculations in the coupled–channel formulation for linearly–polarized light We report the observation of capture–escape resonances in the scattering of electrons from protons in the presence of an intense field. In addition, we investigate the multiphoton ionization of atomic hydrogen in the superintensity range of up to 10^{18} W/cm^2.

2. Formulation

We describe the interaction of an electron with a proton in the presence of an oscillating electric field by the time-dependent(TD) Schrodinger equation in atomic units(a.u.):

$$\left[-\frac{1}{2}\nabla^2 + V(r) + E_0 \cdot r\cos(\omega t)\right]\psi = i\frac{\partial\psi}{\partial t} \quad , \qquad (1)$$

where the Laplacian represents the kinetic energy operator of the electron, V(r) designates the Coulombic interaction $-1/|r|$ of the electron with the proton, and E_0 labels the electric field. We have employed the semiclassical construction in which we treat the particles quantum mechanically and the field classically. In addition, we invoke the usual dipole approximation. For our purposes, the validity of these conventions and approximations is well established. We transform to the Kramers- Henneberger gauge[Gavrila and Kaminski,1984], which we represent schematically by the replacement $r \rightarrow r + \alpha(t)$ with $\alpha(t)$ the classical radius vector of the electron under the influence of only the radiation field. The resulting transformed Schrodinger equation has form

$$\left[-\frac{1}{2}\nabla^2 + V\left[r + \alpha(t)\right]\right]\psi = i\frac{\partial\psi}{\partial t} \quad . \qquad (2)$$

By choosing $A = a\sin\omega t$ for the vector potential of the radiation field, we obtain,

$$\alpha(t) = (a/\omega)\cos\omega t \equiv \alpha_0\cos\omega t, \qquad (3)$$

with the displacement selected along the polar axis($\alpha_0 = \alpha_0\hat{z}$ and $\alpha_0 = E_0/\omega^2$).

We follow the usual Floquet prescription and represent the wavefunction as the product of a periodic component φ and a part depending on a quasienergy E,

$$\psi(r,t) = e^{-iEt}\varphi(r,t) \quad . \qquad (4)$$

We further expand the periodic part in a Fourier series and make a single-center expansion of the spatial function as,

$$\psi(r,t) = e^{-iEt}r^{-1}\sum_{n=-\infty}^{\infty} e^{-in\omega t}\sum_{\ell_n m_n} f_{n\ell_n m_n}(r)Y_{\ell_n m_n}(\hat{r}) \qquad (5)$$

with $\hat{r}$ representing the angular coordinates (θ,ϕ), θ the angle between α_0 and r. Substituting Eq.(5) into (2), multiplying through by a specific Fourier and angular component

$$\exp(in'\omega t)\, Y_{\ell_{n'} m_{n'}}(\hat{r}) \ ,$$

and integrating over a period of the field[$T = 2\pi/\omega$] as well as angle, we derive a set of coupled second order differential equations

$$\left[\frac{d^2}{dr^2} - \frac{\ell_n(\ell_n+1)}{r^2} - k_n^2\right] f_{\ell_n}(r) = \sum_{\ell'} U_{\ell_n,\ell'}(r) f_{\ell'}(r), \qquad (6)$$

where $k_n^2 = (E + n\omega)$,

$$U_{\Gamma',\Gamma}(r) = \sum_{\lambda} \left[\frac{2\ell_n+1}{2\ell_{n'}+1}\right]^{1/2} C(\ell_{n'}\lambda\ell_{n'}|m_n 0 m_n) C(\ell_n \lambda \ell_{n'}|000) v_\lambda^{n'n}(r), \quad (7)$$

and

$$v_\lambda^{n'n}(r) = \left(\frac{2\lambda+1}{\pi}\right) \int_0^\pi dv\ P_\lambda(v) \int_1^1 du\ \frac{V(\vec{r}+\vec{\alpha}_0 u) T_{n'-n}(u)}{[1-u^2]^{1/2}}, \quad (8)$$

with $u \equiv \cos(\omega t)$, $v \equiv \cos(\theta)$, and $T_k(u)$ [$P_k(v)$], the Chebyshev[Legendre] polynomial of order k. The Clebsch–Gordan coefficients are given by $C(l_1 l_2 l_3 | m_1 m_2 m_3)$, and the channels $\Gamma = (n,\ell_n)$ are labeled by a Fourier–state quantum number n and its associated angular momentum quantum number ℓ_n. We extract the scattering information by matching the asymptotic form of these radial components to the usual K–matrix conditions from which we also calculate the S and T matrices. The coupled–channel equations(6) have several important properties: 1) they are block–diagonal in the azimuthal quantum number($m_n = m_{n'} \equiv m$); 2) only channels of the same parity[$(n + \ell_n)$, even or odd] couple; 3), they display a close resemblance to those for electron scattering from a linear ionic diatomic molecule. The first property implies that we may solve the coupled equations independently for each value of m although the Fourier n and partial wave ℓ_n components still couple. The second condition simplifies the form of the structure of the equations by forcing certain matrix elements to vanish. Finally, the last observation has allowed us to bring the full lore of electron–molecule collision theory and computations to bear on the intense field interaction.

In order to solve Eq.(6) numerically, we invoke the close– coupling(CC) approximation by which we truncate the expansion at a finite number of channels,N. In the inner region ($r \leq \alpha_0$), we convert Eq.(6) to a set of coupled integral equations and determine a solution by means of a linear algebraic(LA) prescription(Collins and Schneider,1981; Schneider and Collins,1989). In the outer region, we employ an R–matrix propagation scheme(Light and Walker,1976) to extend the solution into the asymptotic regime. We have utilized the LA method to demonstrate the versatility of our approach. As has been shown for electron–molecule collisions, the LA technique can handle multi–electron systems, nonlocal effects such as exchange and correlation, and the use of different gauges in various spatial regimes(Burke et.al.,1991). The analogy between the scattering equations in the KH gauge and those for electron–molecule collisions can be carried further. The internuclear distance between molecular atoms closely corresponds to the displacement α_0 that represents the separation of effective "charges." The more extended this separation, the larger the basis needed to span the region. We have easily treated displacements of 8 bohr on supercomputers such as a CRAY Y–MP with single symmetries requiring a few minutes of central process time. On further point should be delineated, we recall that the displacement depends on the ratio of the intensity to the square of the frequency. Therefore, for a fixed α_0, we find that the higher the frequency ω, the larger the corresponding intensity that can be handled

The programs have been checked in several ways. First, for the elastic scattering(n n' = 0), we have closed all channels and solved for the bound eigenstates of $U_{nn'}$. For a selection of values of α_0, we obtain excellent agreement with the results of other investigators(Peat et al.,1988, Pont et al.,1990) for the ground σ_g state. Second, we have employed the $e^- + H_2^+$ paradigm to examine an analogous multi state coupling scheme, obtaining agreement for all scattering

quantities with earlier molecular–collision programs(Collins et.al.,1986). We have in all cases sedulously checked the convergence of the scattering quantities as a function of the number of channels(n,ℓ), the number of mesh points, and the matching radius, pushing the convergence to better than a few per cent. For the parameters considered, we generally found that at least a nine–state($n \leq |4|$) CC calculation was required with three to four($\ell \leq 5$ or 7) partial waves for each state.

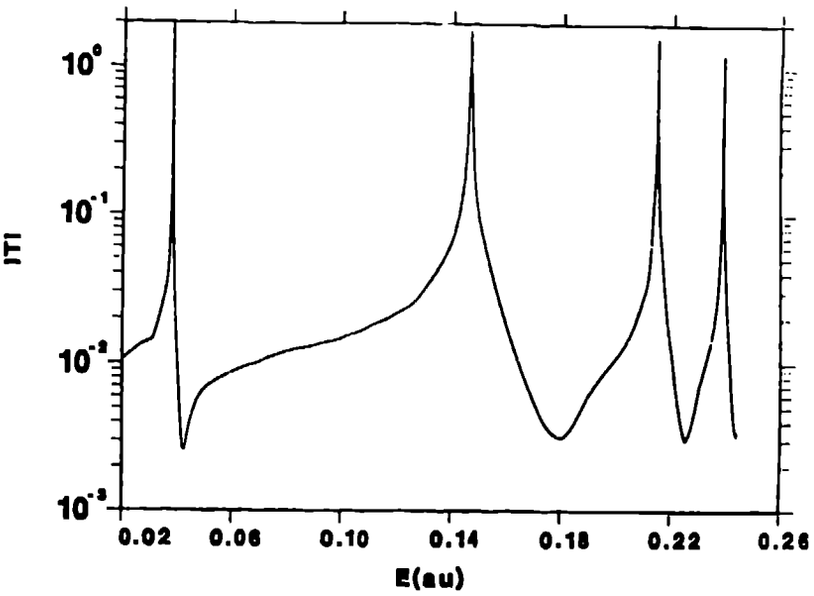


Fig. 1

3. Results and Discussion

a) Electron–Proton Collisions in a Field

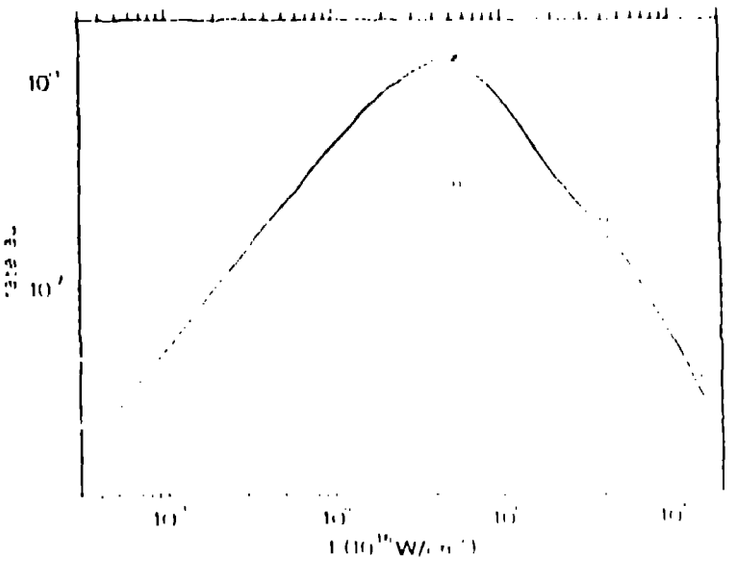
Many interesting phenomena arise when a laser field is introduced into the scattering process. If we examine the cross section or T–matrix elements for $e^- + H^+$ collisons in the absence of an external field, we find a rather mundane functional form without any structural features. However, on allowing the field to participate in the scattering, we observe a number of resonance signatures. These capture–escape resonances, which had been predicted for circularly polarized light[Franz et.al.,1990; Marte and Zoller, 1991], arise form the electron emitting a specific number of photons, becoming temporarily trapped in one of the bound states, and then absorbing an equivalent number of photons to escape. As indicated in Fig.1, we also find these resonances for linearly–polarized light(Collins and Csanak,1991) when the energy difference between the electron and n–photons approximately equals the binding energy of a bound state. We display the magnitude of the elastic scattering T–matrix element[(0,2) → (0,2)] as a function of the quasienergy for an electric field E_0 of 0.0207au($I=1.5\times10^{13}$ W/cm^2) and a frequency of ω 0.27 hartrees(7.35eV) with m 0 and even parity[α_0 0.284 bohr]. The lowest peak corresponds to the two photon capture into the ground 1s state of H. From simple energy conservation arguments, we expected the position to be 0.04h. However, our eleven state CC calculation places the resonance at a lower energy of 0.0376h. This shift demonstrates the great importance of channel coupling and large CC bases in the proper representation of resonance phenomena since for the five state case($n \leq |2|$), the lowest coupling order at which double photon processes can appear, we observed an upward shift. As more states are included($N > 11$), the position moves below 0.04h and converges on the final value given above. The remaining series of resonances is associated with single photon transitions to the excited states of H with principal quantum numbers equal to 2,3,and 4 respectively. The positions of these resonances shift

to higher energies than expected from pure energy conservation considerations, indicating the strong interaction among the various channels. This series, of course, converges to the ionization limit at 0.27h and forms an ideal candidate for a MCQD approach suggested by Marte and Zoller(1991). We have found two interesting trends. If we fix the frequency and increase the intensity, we observe the resonance position moves to lower energies and the width broadens. Conversely, if we set the intensity and increase the frequency, we note a decrease in width and upward shift in the position. These observations apply only to regimes governed by the same order of multiphoton process. For example, by increasing the intensity, we eventually encounter a situation in which the sum of the quiver($E_0^2/4\omega^2$) and binding energies exceeds the photon energy($n\omega$). In order to promote the electron into the field, we must invoke a higher-order photon process(n+1). For this situation, the trends may be interrupted or reversed depending on the strength of the coupling.

b) **Multiphoton Ionization of Hydrogen**

We have also performed calculations of the ionization rates for the multiphoton ionization from atomic hydrogen. We identify the ionization rate with the width of the resonance corresponding to the desired initial state, which is permitted if the photon energy is significantly larger than the ionization potential(Alber and Zoller,1988; Kyrola, 1986). We determine the width from a fit of the eigenphase sum as a function of quasienergy using the Breit-Wigner form. For various frequencies and intensities, ranging from 10^{12} to 10^{18} W/cm^2, we obtain excellent agreement with other Floquet(Pont et.al.,1991) and TD(Kulander,1987; Dorr et.al.,1991) calculations for ground-state ionization. As a representative calculation, we explore the superintense regime for single-photon ionization(ω = 1.au) for H(1s). In Fig. 2, we display the ionization rate as a function of intensity for an 15-state CC calculations with five partial waves per state(N=75). We find that as the intensity increases, the rate at first follows suit. However, eventually the rate turns over and decreases for a superintense field. This characterizes the "stabilization" or ionization suppression case(Su and Eberly,1991; M.Dorr et.al.,1991; Kulander et.al.,1991) seen in TD and Floquet models. The electron becomes partially trapped near the nucleus at the higher intensities thus lowering the probability of escape. We compare against TD calculations(Kulander et.al.,1991), represented by the open circles, and find excellent agreement at the very high field strengths. Around the rate maximum, the agreement is poorer possibly due to greater sensitivity to the manner in which the field is ramped.

Fig. 2



4. Summary

We have extended the approach of Gavrila and Kaminski to the solution of the Schrodinger equation in the Kramers–Henneberger gauge for electron–proton scattering and photoionization in an intense linearly–polarized radiation field using a Floquet–Fourier expansion and solving the resulting set of coupled differential equations. We observe the one– and two–photon capture–escape resonances and examined their behavior as a function of field strength. In addition, we find evidence of the ionization suppression mechanism for very high intensities.

We wish to acknowledge useful discussions with Dr. A.L.Merts and Dr. P. Milonni. Work performed under the auspices of the U.S. Department of Energy through the Theoretical Division of the Los Alamos National Laboratory.

References

Alber F. and P. Zoller, Phys. Rev. A **37**, 377(1988).
Bhatt R., B. Piraux, and K. Burnett, Phys. Rev. A **37**, 98 (1988).
Burke P.G., P. Francken, and C.J. Joachain, J. Phys. B **24**, 761 (1991).
Burnett K., P.L. Knight, B.R.M. Pireux, and V.C. Reed, Phys.Rev.Lett. **66**.301(1991).
Collins L.A. and G. Csanak, Phys.Rev.A,**44**, R5343(1991).
Collins L.A. and B.I. Schneider, Phys. Rev. A **24**, 2387 (1981).
Collins L.A., B.I. Schneider, C.J. Noble, C.W. McCurdy, and S. Yabushita, Phys. Rev. Lett. **57**, 980 (1986).
Dimou L. and F.H.M. Faisal, Phys. Rev. Lett. **59**, 872 (1987).
Dorr M, R.M. Potvliege, D. Proulx, and R. Shakeshaft, Phys.Rev.A **43**, 3729(1991).
Faisal F.H.M., J. Phys. B **6**, L89 (1973).
Franz A., H. Klar, J.T. Broad, and J.S. Briggs, J. Opt. Soc. Am. B **7**, 545 (1990).
Gavriia M. and J.Z. Kaminski, Phys. Rev. Letts. **52**, 613 (1984).
Gersten J.I., and M.H. Mittleman J. Phys. B **9**, 25561 (1976).
Giusti–Suzor A. and P. Zoller, Phys. Rev. A **36**, 5178 (1987).
Henneberger W.C., Phys. Rev. Letts, **21**, 838 (1968).
Kramers H.A., Collected Scientific Papers (North Holland, Amsterdam, 1956), p. 262.
Kulander, K., Phys. Rev. A **35**, 445(1987).
Kulander, K., K.T. Schafer, and J.L. Krause, Phys. Rev. Lett. **66**, 2601(1991).
Kyrola K., J. Phys. B **19**, 1437(1986).
Marte P. and P. Zoller, Phys. Rev. A **43**, 1512 (1991).
Light J.C. and R.B. Walker, J. Chem. Phys. **65**, 4272 (1976).
Pont M., N.R. Walet, M. Gavrila, and C.W. McCurdy, Phys. Rev. Letts. **61**, 939 (1988).
Pont M., Phys. Rev. A **40**, 5659 (1989).
Pont M. and M. Gavrila, Phys. Rev. Letts, **65**, 2362 (1990).
Pont M., N. Walet, and M. Gavrila, Phys. Rev. A **41**,477(1990).
Pont, M. D. Proulx, and R. Shakeshaft, Phys.Rev. A **44**, 4486(1991)
Potvliege, R.M. and R. Shakeshaft, Phys. Rev. A **40**, 3061(1989).
Schneider B.I. and L.A. Collins, Comp. Phys. Rep. **10**, 49 (1989).
Su Q. and J.H.Eberly, Phys. Rev. A **43**, 2474(1991).